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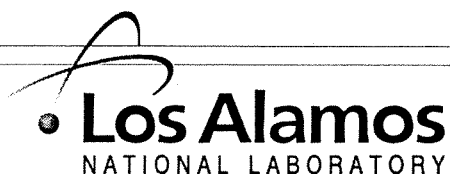
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Title: A CORRELATION FOR STEEL CORROSION IN  
NON-ISOTHERMAL LBE LOOPS

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## ORIGINAL PAPER

A Correlation for Steel Corrosion in Non-isothermal LBE Loops<sup>1</sup>

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The mass transfer controlled corrosion of steels in non-isothermal LBE loop system was studied based on a kinetic corrosion model. Variations of Sherwood number at the highest temperature isothermal leg in DELTA loop (a non-isothermal Lead-Bismuth Eutectic (LBE) test loop set up at the Los Alamos National Laboratory) were examined to find out the dependence of the average mass transfer at the highest temperature leg on the axial conditions. The average Sherwood number at the highest temperature leg (test leg) is correlated by a dimensionless equation:

$$Sh_{av} = 0.53Re^{0.6}Sc^{1/3}(d/L)^{1/3}(\Delta T/T_{\max})^{1/3},$$

where  $Re$  and  $Sc$  are Reynolds number and Schmidt number, respectively,  $d$  (m) is the tube diameter and  $L$  (m) is the loop length,  $\Delta T$  (°C) is the temperature difference between the highest and the lowest temperature and  $T_{\max}$  (°C) is the maximal temperature.

**KEYWORDS:** corrosion, mass transfer, lead bismuth, non-isothermal loop

## I. Introduction

For practical evaluation of the corrosion rate of steels exposed to flowing lead-bismuth eutectic (LBE) with oxygen control, a non-isothermal material test loop named DELTA loop has been built at the Los Alamos National Laboratory (LANL).<sup>1)</sup> It has been reported elsewhere that the DELTA is being prepared for corrosion testing of kinds of materials in LBE as a part of the Advanced Fuel Cycle Initiative program<sup>2)</sup> (AFCI, formerly the Advanced Accelerator Applications (AAA) Program).

In recent years, investigations on the corrosion of materials exposed to LBE with or without oxygen control have been carried out experimentally<sup>3–6)</sup> and theoretically.<sup>7–10)</sup> Previous results indicate that the corrosion in a non-isothermal LBE system depends on many factors: time of exposure, thermal gradient, LBE flow velocity, solid and liquid compositions, etc., and occurs through several mechanisms: dissolution, erosion, mass transport, phase transport, etc.<sup>7)</sup>

The present study focuses on the mass transfer controlled corrosion. The corrosion product concentration (mainly iron) in the bulk LBE is derived by solving the convective transport equation in the bulk based on a kinetic corrosion model.<sup>8,9)</sup> The iron concentration in the bulk LBE varies along the axis and the variation is small enough to be neglected in DELTA loop. Comparisons between Sherwood number at the highest temperature leg in DELTA loop and that in an isothermal open pipe system with the same conditions, illustrate that the Sherwood number at the highest temperature leg in a non-isothermal loop depends on loop temperature differences. A correlation for the average Sherwood number at the highest temperature isothermal leg in a non-isothermal LBE loop is derived through analyzing the corrosion rate at

the highest temperature leg in DELTA loop. The correlation is benchmarked by application to a non-isothermal liquid lead loop<sup>11)</sup> for which experimental results are available. Additionally, the correlation can be used easily to calculate the average corrosion rate at the highest temperature leg and explicitly shows the dependence of corrosion on the temperature difference and the loop length.

## II. Corrosion Model for Non-isotherm Liquid Metal Loops

By solving the mass transport equation in the sub-laminar boundary layer, previous studies<sup>8,9)</sup> obtained the solution of the corrosion/precipitation flux  $q$  from/to the tube wall for a non-isothermal liquid metal loop:

$$q(\xi) = -D \frac{\partial c}{\partial y} \Big|_{y=0} = \left( \frac{2\pi D^2 \gamma}{3L} \right)^{1/3} \times \frac{1}{Ai(0)\Gamma(1/3)} \sum_k Q_k \exp(2\pi k i \xi), \quad (1)$$

where  $\xi=x/L$  is the non-dimensional coordinate in the axial direction,  $y$  denotes the transverse coordinate with  $y=0$  at the tube surface,  $Ai$  and  $\Gamma$  are Airy Function and Gamma Function, respectively,  $\gamma$  is the wall shear rate defined by  $\gamma=\lambda V^2/2\nu$  ( $\lambda$  is the friction factor,  $V$  (m/s) is the bulk velocity,  $\nu$  (m<sup>2</sup>/s) is the kinematic viscosity),  $D$  (m<sup>2</sup>/s) is the diffusion coefficient of the corrosion product considered and  $Q_0=0$ ,  $Q_k=a_k k^{1/3} i^{1/3}$  for  $k>0$  and  $Q_k=a_k |k|^{1/3} (-i)^{1/3}$  for  $k<0$ .  $i$  is the imaginary unit, and it is chosen<sup>9)</sup>  $i^{1/3}=\sqrt{3}/2+i/2$  and  $(-i)^{1/3}=\sqrt{3}/2-i/2$ . The surface corrosion product concentration is assumed to be a function of the axial coordinate and it can be expanded into the following Fourier series:

$$c_w(\xi) = \sum_k a_k \exp(2\pi k i \xi). \quad (2)$$

For an LBE system, the corrosion of structural materials,

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steels in particular, usually occurs in two different ways: reduction and dissolution,<sup>8)</sup> depending on the oxygen concentration in LBE. No matter in which way the corrosion occurs, it is usually sufficiently fast that corrosion product concentrations are always at their saturated or equilibrium levels. Reference 8) suggested the surface concentration of Fe (main corrosion product) in an oxygen controlled LBE system could be calculated through the following equation:

$$c_{Fe} = \text{Min}(10^{6.01-4.380/T}, c_O^{-4/3} 10^{11.35-(12.844/T)}), \quad (3)$$

where  $c_{Fe}$  (ppm) and  $c_O$  (ppm) are concentration of iron at the tube surface and concentration of oxygen in LBE, respectively,  $T$  (K) is the local temperature. Therefore, if the axial temperature profile is given, the corrosion and precipitation rates and profiles can be obtained through Eqs. (1)–(3).

### III. Bulk Concentration Distributions

For an open pipe flow, the corrosion product concentration in the bulk may be assumed to be zero or equals to that at the inlet because new fluids continuously flow into the pipe at the inlet. For a closed loop flow, the fluid is not renewed, and the bulk corrosion product concentration increases in time until it reaches a steady state. This transient time can be very short if the test loop is constructed of materials with corrosion resistance similar to that of the test materials, as is the case for the majority of LBE test loops. At steady state, the convective mass transfer process in the bulk flow satisfies the following equation:

$$AV \frac{dc_b}{dx} = pq[\xi(x)], \quad (4)$$

where  $A$  (m<sup>2</sup>) and  $p$  (m) are the area and the perimeter of the cross-section of the circulate tube. For the circular tube  $A=\pi d^2/4$  and  $p=\pi d$ . Regarding that  $c_b$  is real, integrate the above equation, we get:

$$c_b(\xi) = c_b^0 + \frac{P}{AV} \frac{(2\pi L^2 D^2 \gamma)^{1/3}}{3^{1/3} \text{Ai}(0) \Gamma(1/3)} \sum_k P_k \exp(2\pi k i \xi), \quad (5)$$

where  $P_0=0$ ,  $P_k=Q_k/(2\pi k i)$  for  $k>0$  and  $P_k=\bar{P}_{-k}$  for  $k<0$  (the bar presents the conjugate value),  $c_b^0$  is the average bulk concentration, for a simple loop, it equals to the average surface species concentration  $a_0$ .<sup>9)</sup>

Equation (5) is used to calculate the bulk concentration of iron in DELTA loop. The loop length is 29.92 m and the tube diameter is 0.0525 m. Detailed description of the loop can be found in Ref. 1). The axial temperature profile is shown in Fig. 1 and the calculated iron bulk concentration distributions for different velocities along the axis are shown in Fig. 2. The diffusion coefficient and the kinematic viscosity of LBE are chosen as  $D=10^{-9}$  m<sup>2</sup>/s and  $\nu=1.5 \times 10^{-7}$  m<sup>2</sup>/s that are from Ref. 8). The Blasius expression for the friction factor is used in all the calculations in this paper.

Figure 2 shows that the iron bulk concentration is not zero. It varies along the axis and reaches the highest value at the end of the test leg (the highest temperature leg). The variation becomes smaller with the bulk velocity increasing.

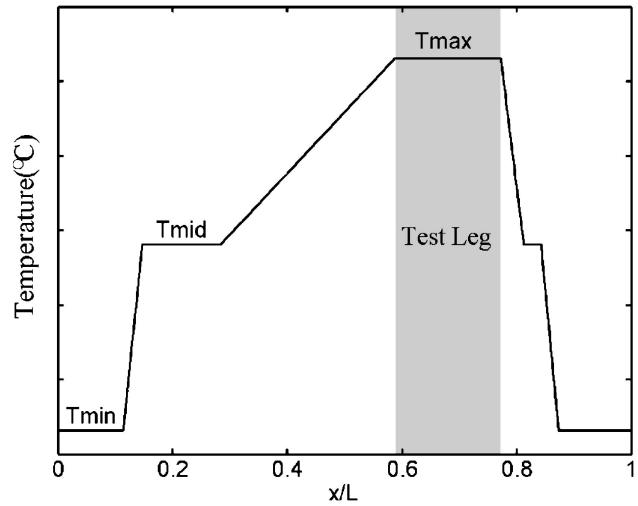


Fig. 1 Temperature profile of the DELTA loop,  $T_{max}$ : maximal temperature;  $T_{min}$ : minimal temperature;  $T_{mid}$  middle temperature.  $T_{mid}=T_{min}+\Delta T/2$  and  $\Delta T=T_{max}-T_{min}$

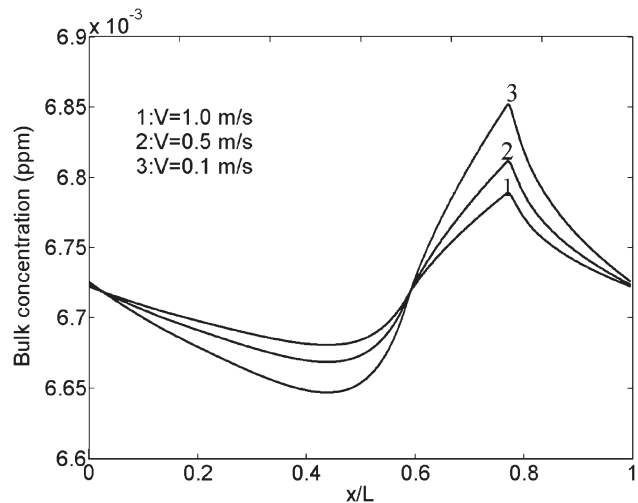


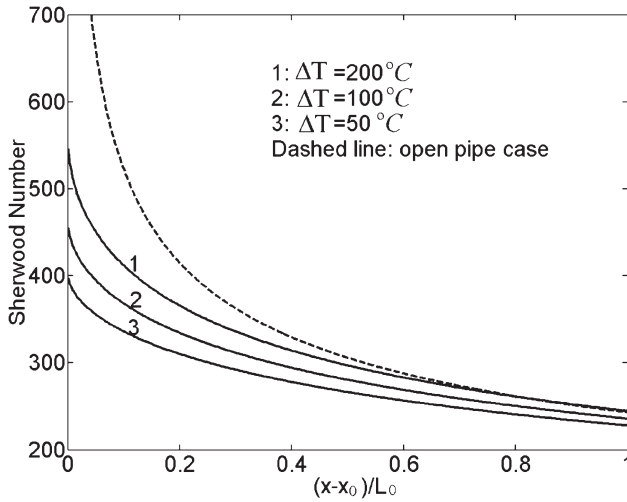
Fig. 2 Fe bulk concentration profile along loop axis in DELTA loop for different flow velocities,  $T_{max}=550^\circ\text{C}$ ,  $\Delta T=200^\circ\text{C}$  and  $c_O=0.01$  ppm

However, even for a small bulk velocity, the variation is small enough to be neglected. For example, the difference between the maximum and the minimum bulk concentrations for  $V=0.1$  m/s is less than 3% of the average bulk concentration. Therefore, it is reasonable to assume the bulk concentration to be constant along the axis at the steady state for DELTA loop.

### IV. Variations of Sherwood Number in the Test Leg of DELTA Loop

In the mass transfer controlled corrosion, the corrosion rate can also be expressed in a dimensionless form by Sherwood number that is defined by:<sup>7)</sup>

$$Sh = \frac{Kd}{D}, \quad (6)$$



**Fig. 3** Sherwood number variations in the highest temperature test leg in DELTA loop,  $x_0$ : the beginning coordinate of the test leg,  $L_0$ : the test leg length.  $T_{\max}=550^\circ\text{C}$ ,  $c_0=0.01\text{ppm}$  and  $V=0.5\text{ m/s}$

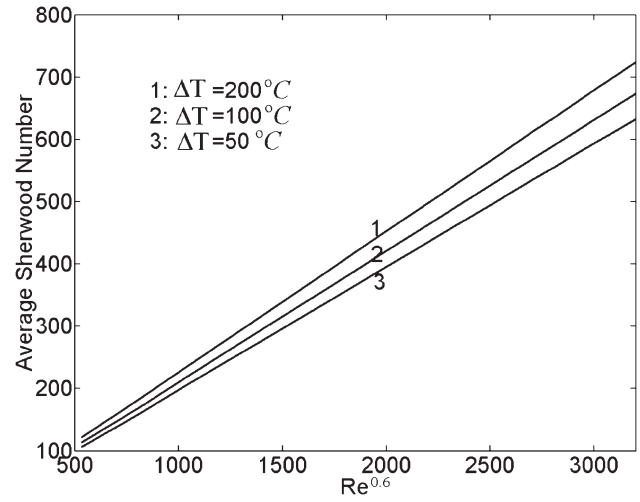
where  $K$  (m/s) is the mass transfer coefficient and is calculated by

$$K(\xi) = \frac{q(\xi)}{c_w(\xi) - c_b(\xi)}. \quad (7)$$

In our previous studies,<sup>8,9)</sup> we have analyzed the corrosion/precipitation profiles in DELTA loop, which indicates that the corrosion at the highest temperature leg depends strongly on axial temperature differences. Experimental results from one non-isothermal loop cannot be directly applied to another loop with different temperature profile. In the present study, we will correlate the corrosion rate at the highest temperature leg in a non-isothermal liquid metal loop with the temperature difference as well as the maximal temperature through analyzing variations of Sherwood number at the highest temperature leg in DELTA loop.

The mass transfer variation at the highest temperature leg (test leg) in DELTA loop, expressed as the Sherwood number, is shown in **Fig. 3**. For comparisons, the Sherwood number for an open pipe flow obtained from an analytical solution<sup>12)</sup> is also shown in the figure. The figure shows that the mass transfer decreases sharply with increasing the distance from the beginning of the highest temperature leg. The mass transfer for DELTA loop at the highest temperature is less than that for an open pipe flow with the same conditions. It also depends on the temperature difference between the maximal temperature and the minimal temperature  $\Delta T$ . Therefore the effects of the temperature difference have to be regarded when correlating the Sherwood number with the thermal hydraulic parameters such as the Reynolds number and the Schmidt number for cases of non-isothermal loop.

To capture the mass transfer characteristics at the isothermal leg in a non-isothermal loop, the average Sherwood number  $Sh_{av}$  at the highest temperature leg is examined. It can be shown by dimensional analysis that  $Sh_{av}$  number is a function of the Reynolds number ( $Re=Vd/\nu$ ) and Schmidt



**Fig. 4** The average Sherwood number vs.  $Re^{0.6}$  in the test leg of the DELTA loop,  $T_{\max}=550^\circ\text{C}$ ,  $c_0=0.01\text{ ppm}$

number ( $Sc=\nu/D$ ). For the open pipe flow case, the relation can be theoretically obtained.<sup>7)</sup> For a non-isothermal loop, the Sherwood number should also be a function of the global conditions. It is assumed:

$$Sh_{av} = b_0 Re^{b_1} Sc^{b_2} (d/L)^{b_3} (\Delta T/T_{\max})^{b_4}. \quad (8)$$

Theoretical analyses<sup>9)</sup> show that  $b_1=0.6$  and  $b_2=b_3=1/3$  for high  $Sc$  number when the mass transfer process is controlled by mass transfer through the sub-laminar layer. **Figure 4** shows variations of the average Sherwood number as a function of  $Re^{0.6}$ . The figure clearly shows that the Sherwood number scales as  $Re^{0.6}$ . Because the temperature profile is a global condition similar to the loop length, we assume  $b_4=1/3$  like the dependence on the loop length. The variations of the average Sherwood number as functions of  $\Delta T^{1/3}$  and  $T_{\max}^{-1/3}$  are shown in **Figs. 5(a)** and **(b)**. The figure indicates that the average number scales as  $\Delta T^{1/3}$  and  $T_{\max}^{-1/3}$  in the range if  $50^\circ\text{C} \leq \Delta T \leq 200^\circ\text{C}$  and  $450^\circ\text{C} \leq T \leq 650^\circ\text{C}$ . Therefore  $b_4=1/3$ .

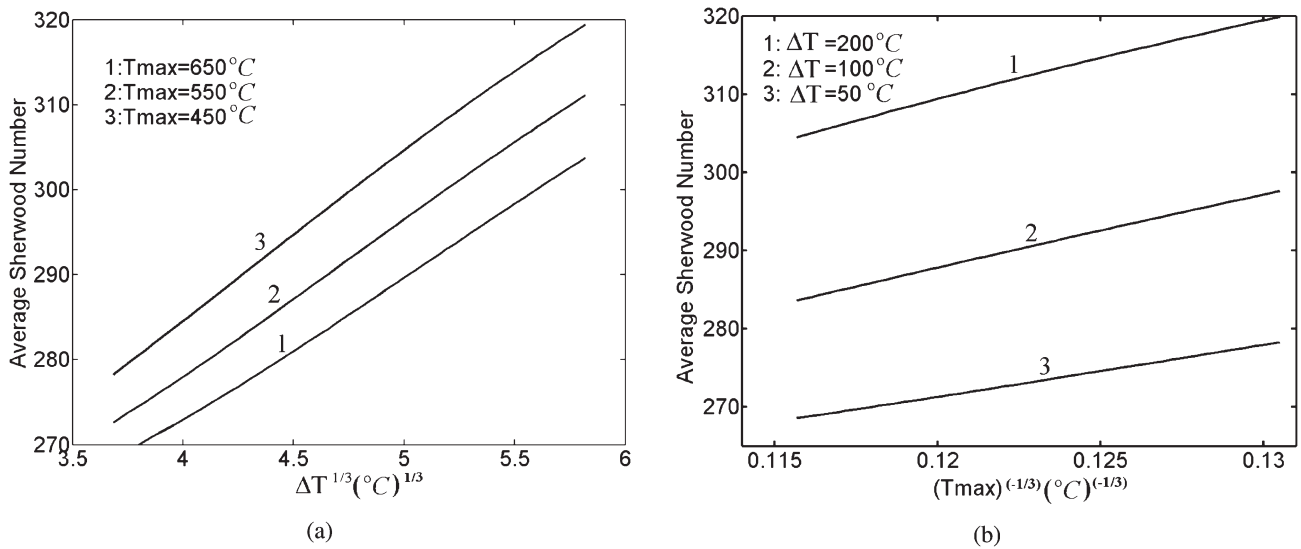
To obtain the value of  $b_0$ , the variation of the Sherwood number as a function of  $Re^{0.6} Sc^{1/3} (d/L)^{1/3} (\Delta T/T_{\max})^{1/3}$  is shown in **Fig. 6** for three operation conditions in the range  $50^\circ\text{C} \leq \Delta T \leq 200^\circ\text{C}$  and  $0.1\text{ m/s} \leq V \leq 2.0\text{ m/s}$ . The fitted value of  $b_0$  is 0.53. The results obtained from Eq. (8) using  $b_0=0.53$  agree very well with the results from the kinetic model. Therefore, the Sherwood number at the highest temperature leg in DELTA loop, in the ranges of  $0.1\text{ m/s} \leq V \leq 2.0\text{ m/s}$ ,  $50^\circ\text{C} \leq \Delta T \leq 200^\circ\text{C}$  and  $450^\circ\text{C} \leq T_{\max} \leq 650^\circ\text{C}$ , is correlated by:

$$Sh_{av} = 0.53 Re^{0.6} Sc^{1/3} (d/L)^{1/3} (\Delta T/T_{\max})^{1/3}. \quad (9)$$

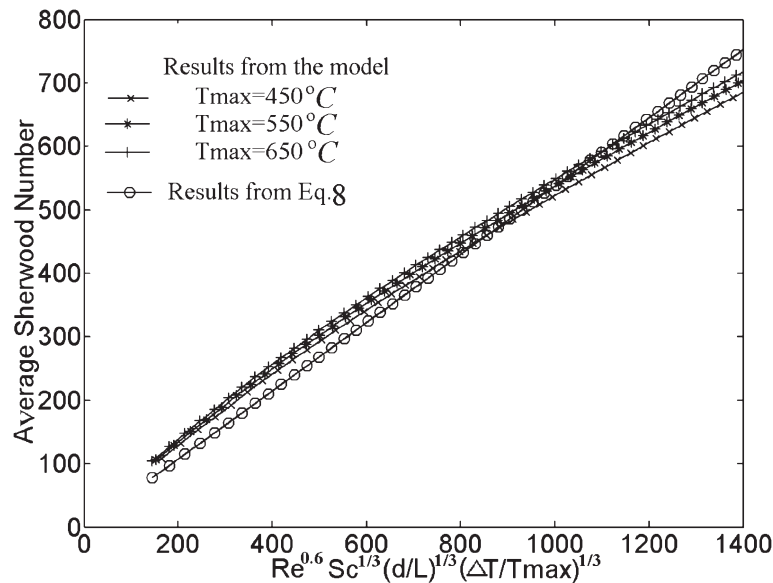
Equation (9) explicitly shows the relation of the mass transfer in the highest temperature leg in a non-isothermal liquid metal loop with the temperature difference and the whole loop length.

## V. Application to Experiments in a Pure Lead Loop

Although the correlation is obtained by analyzing the



**Fig. 5** (a) The average Sherwood number vs.  $\Delta T^{1/3}$  in the test leg of the DELTA loop (b) The average Sherwood number vs.  $T_{\text{max}}^{-1/3}$  in the test leg of the DELTA loop,  $V=0.5$  m/s,  $c_0=0.01$  ppm



**Fig. 6** Comparisons between the results from the model and that from Eq. (8)

mass transfer characteristics in DELTA loop, it is expected to be suitable for other non-isothermal liquid metal loops in which the surface corrosion product concentration can be written into a similar expression to Eq. (3). To benchmark the correlation, it is applied to a non-isothermal pure lead loop built by Sanier and Santarini.<sup>11)</sup> The temperature profile for the pure lead loop is shown in **Fig. 7** in a solid line. Experiments were carried out using pure lead without oxygen control (it is assumed to be in oxygen-depleted state by use of a reducing cover gas). Experiments are carried out for two types of steels at the highest temperature leg. For steel 10 CD 9–10, the experimental corrosion rate is between  $75 \pm 20$ – $110 \pm 20$   $\mu\text{m}$  after 3,000 h and for steel Z 10 CD Nb V 92 is between  $25 \pm 20$ – $40 \pm 20$   $\mu\text{m}$  after 2,800 h.

To calculate the corrosion rate at the test leg in the pure

lead loop, the surface iron concentration is calculated by<sup>13)</sup>  $c_{\text{Fe}} (\text{ppm}) = 10^{4.34 - 3450/T(\text{K})}$ . The following parameters are considered: maximal temperature:  $T_{\text{max}}=550^{\circ}\text{C}$ , temperature difference:  $\Delta T=85^{\circ}\text{C}$ , flow velocity:  $V=0.115$  m/s, loop length:  $L=6$  m, internal diameter of the pipe:  $d=0.0247$  m, and the viscosity:<sup>7)</sup>  $\nu=1.65 \times 10^{-7}$   $\text{m}^2/\text{s}$ . For the diffusion coefficient  $D$ , we employ the equation developed by Robertson,<sup>14)</sup>  $D (\text{m}^2/\text{s}) = 4.9 \times 10^{-7} \exp[-Q/RT (\text{K})]$ , where  $R$  is the gas constant and  $Q=44,100 \pm 6,300$  J/mol. Therefore at  $550^{\circ}\text{C}$ , we get  $D=0.31$ – $2.0 \times 10^{-9}$   $\text{m}^2/\text{s}$ . Then we get  $Sh_{\text{av}}=69.6$ – $128.5$  at the highest temperature leg. The calculated bulk concentration iron is shown in **Fig. 7** (dashed line) and it is found  $c_b^0=0.8555$  ppm. The corrosion rate is calculated by:

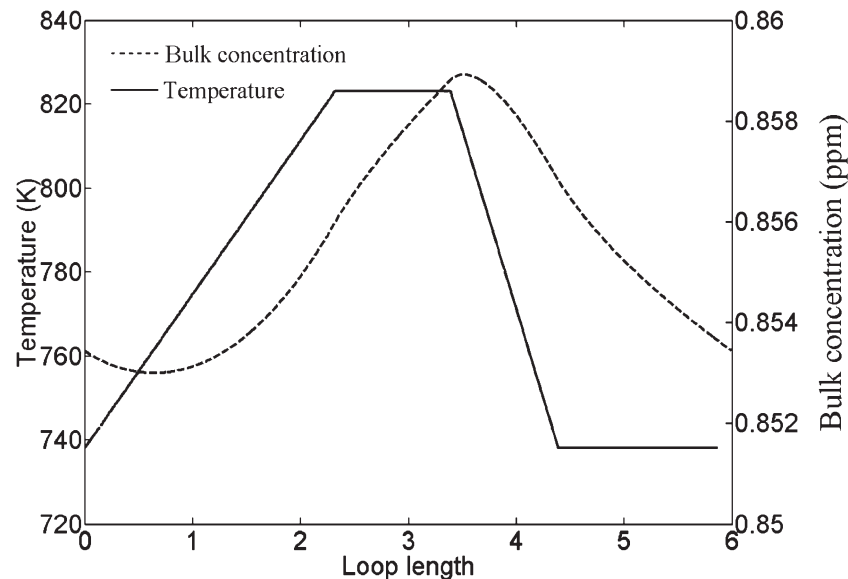


Fig. 7 Temperature profile of the pure lead loop<sup>11)</sup> and the calculated bulk iron concentration

$$J = K(c_w - c_b^0) = Sh_{av}D/d(c_w - c_b^0),$$

and we get the iron corrosion rate is 13–44  $\mu\text{m}$  per 3,000 h. Clearly, the calculated results from the correlation (Eq. (9)) are sensitive to the diffusion coefficient  $D$ . There are some deviations between the calculated and experimental results, but these deviations are probably expected due to experimental uncertainties, the value of the diffusion coefficient, the alloy composition effects and other corrosion mechanisms such as erosion-corrosion. On the other hand, the local corrosion rate can be estimated directly from Eq. (1). Employing  $D=2.0 \times 10^{-9} \text{ m}^2/\text{s}$ , our previous studies<sup>9)</sup> predict a corrosion rate at the high temperature leg of the pure lead loop is between 40–70  $\mu\text{m}$  per 3,000 h, which is also close to the experimental results.

## VI. Conclusions

Corrosion product concentrations in the bulk flow in a non-isothermal closed loop were examined. A correlation for Sherwood number with system conditions was developed based on analyzing the mass transfer characteristics at the test leg of DELTA loop using a kinetic corrosion model. The correlation is partially validated by application to experimental results from a pure lead loop. The correlation indicates that the average corrosion rate at the highest isothermal leg in a non-isothermal liquid metal loop is proportional to  $(\Delta T/T_{\max}L)^{1/3}$  that are determined by the axial loop conditions. The present calculations give a very preliminary dependence relation between corrosion rates and the global conditions of a non-isothermal loop. Systematical (both experimental and theoretical) studies are needed to determine corrosion behaviors of practical LBE nuclear systems.

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